Induced polarization of clay-sand mixtures: experiments and modelling

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Frequency-domain induced polarization (IP) measurements consist of imposing an alternative sinusoidal electrical current (AC) at a given frequency and measuring the resulting electrical potential difference between two other non-polarizing electrodes. The magnitude of the conductivity and the phase lag between the current and the difference of potential can be expressed into a complex conductivity with the in-phase representing electromigration and a quadrature conductivity representing the reversible storage of electrical charges (capacitive effect) of the porous material. Induced polarization has become an increasingly popular geophysical method for hydrogeological and environmental applications [1]. These applications include for instance the characterization of clay materials used as permeability barriers in landfills or to contain various types of contaminants including radioactive wastes [2]. The goal of our study is to get a better understanding of the influence of the clay content, clay mineralogy, and pore water salinity upon complex conductivity measurements of saturated clay-sand mixtures in the frequency range ~ 1 mHz-12 kHz.

The complex conductivity of saturated unconsolidated sand-clay mixtures was experimentally investigated using two types of clay minerals, kaolinite and smectite in the frequency range 1.4 mHz - 12 kHz. Four different types of sample were used, two containing mainly kaolinite (80% of the mass, the remaining containing 15% of smectite and 5% of illite/muscovite; 95% of kaolinite, 5% of illite/muscovite), and the two others containing mainly Na-smectite or Na-Ca-smectite (95% of the mass; bentonite). The experiments were performed with various clay contents (1, 5, 20, and 100 % in volume of the sand-clay mixture) and salinities (distilled water, 0.1 g/L, 1 g/L, and 10 g/L NaCl solution). In total, 44 saturated clay or clay-sand mixtures were prepared. Induced polarization measurements were performed with a cylindrical four-electrode sample-holder (cylinder made of PVC with 30 cm in length and 19 cm in diameter) associated with a SIP-Fuchs II impedance meter and non-polarizing $Cu/CuSO_4$ electrodes (Figure 1). These electrodes were installed at 10 cm from the base of the sample holder and regularly spaced (each 90 degree).

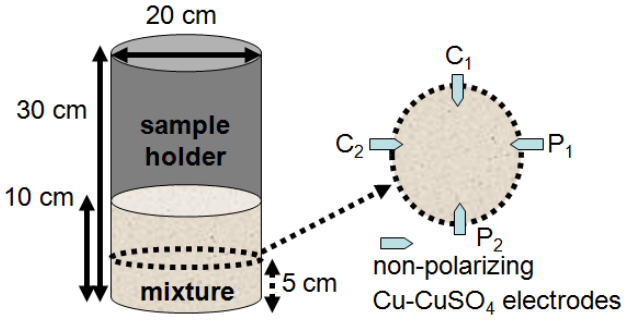

Figure 1: Illustration of the sample holder dimensions used for the measurement

The results illustrate the strong impact of the Cationic Exchange Capacity (CEC) of the clay minerals upon the complex conductivity. The amplitude of the in-phase conductivity of the kaolinite-clay samples is strongly dependent to saturating fluid salinity (Figure 2) for all volumetric clay fractions, whereas the in-phase conductivity of the smectite-clay samples is quite independent on the salinity, except at the low clay content (5% and 1% of clay in volume). This is due to the strong and constant surface conductivity of smectite associated with its very high CEC. The quadrature conductivity increases steadily with the CEC and the clay content. We observe that the dependence on frequency of the quadrature conductivity of sand-kaolinite mixtures is more important than for sand-bentonite mixtures. For both types of clay, the quadrature conductivity seems to be fairly independent on the pore fluid salinity (Figure 2) except at very low clay contents (1% kaolinite-clay in volume). This is due to the constant surface site density of Na counter-ions in the Stern layer of clay materials [3]. At the lowest clay content (1%), the magnitude of the quadrature conductivity increases with the salinity, as expected for silica sands. In this case, the surface site density of Na counter-ions in the Stern layer increases with salinity [4].

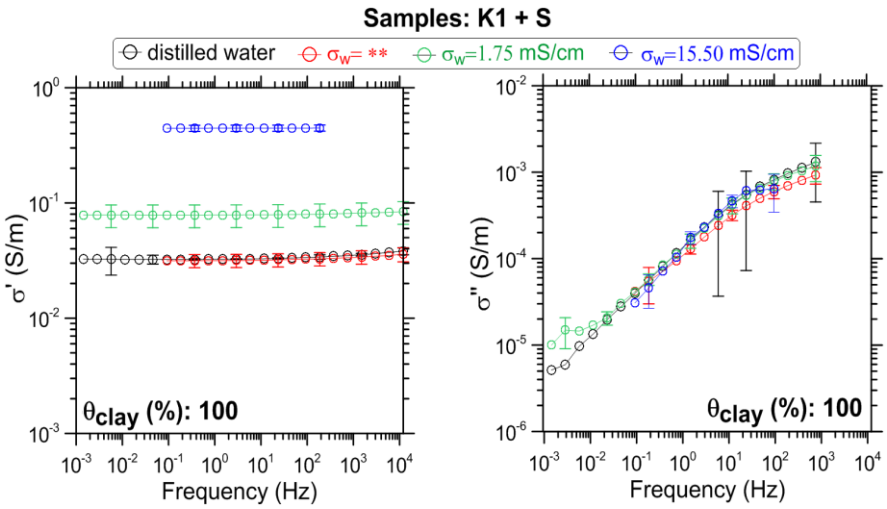

Figure 2: In-phase and quadrature conductivities of "kaolinite" clay for different fluid salinities

The experimental data show good agreement with predicted values given by our Spectral Induced Polarization (SIP) model [4]. This complex conductivity model considers the electrochemical polarization of the Stern layer coating the clay particles and the Maxwell-Wagner polarization. We use the differential effective medium theory to calculate the complex conductivity of the porous medium constituted of the grains and the electrolyte. The SIP model includes also the effect of the grain size distribution upon the complex conductivity spectra. Interfacial parameters are estimated using the TLM of Sverjensky [5] for silica and the Donnan model of Tournassat and Appelo [6] for smectite.

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